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POLYACETYLENE AND RELATIVISTIC FIELD-THEORY MODELS

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Connections between continuum, mean-field, adiabatic Peierls-Fröhlich theory in the half-filled band limit and known field theory results are discussed. Particular attention is given to the ϕ^4 model and to the solvable N = 2 Gross-Neveu model. The latter is equivalent to the Peierls system at a static, semi-classical level. Based on this equivalence we note the prediction of both kink and polaron solitons in models of trans-$(CH)_x$. Polarons in cis-$(CH)_x$ are compared with those in the trans isomer. Optical absorption from polarons is described, and general experimental consequences of polarons in $(CH)_x$ and other conjugated polymers is discussed.

1. INTRODUCTION.

In recent months there has been an enthusiastic and growing appreciation of relationships between models of conjugated polymers such as polyacetylene ($(CH)_x$) and relativistic field theory models. A notable example is the feature of fractionally charged defect states with unusual spin/charge assignments.[1] This has a several year history in field theory and Peierls-Fröhlich charge density wave theory. The topic is covered in detail by Schrieffer,[1] so we will not dwell on it here. Rather, in this paper we comment on two general areas of connections between model field theories and half-filled band Peierls systems, which have augmented existing theoretical descriptions of polyacetylene:[2]

(a) Equivalences between field equations. The level of equivalence has to be made explicit in individual cases. Here we will use an exact equivalence[2] between the static, semiclassical N = 2 Gross-Neveu (G-N) field theory[3] and the continuum limit[4] of the half-filled band Peierls chain, described in a discrete Hückel basis by Su et. al.[1,5] and solved in a mean-field, adiabatic approximation: for brevity we will refer to the continuum theory as the TLM equa-

tions.[4] It is not appropriate here to describe the G-N model in detail (see ref. 2 for a complete description and proof of the equivalence). We remark only that the general N-component G-N model[3] is a one-space, one-time dimensional field theory of self-coupled fermions of N "types": our equivalence uses N = 2, corresponding to the spin degeneracy. G-N is of interest to field theorists as a solvable model[6] exemplifying a number of crucial concepts developed by them in recent years; asymptotic freedom, dynamical spontaneous symmetry-breaking, ultraviolet renormalization, dimensional transmutation, charge conjugation symmetry, etc. It is certainly not appropriate to explain these terms here. (See Ref. 2.) It is enough to note that they all have precise analogues in the TLM equations, which explains the field theorists' enthusiasm[1] for $(CH)_x$! The specific converse value to $(CH)_x$ theorists is the prescription of a constructive procedure[2] for determining all static solutions of the TLM equations: this follows because of the truly soliton[6] nature of the models which has already been fully exploited in field theory literature.[2,3]

(b) Qualitative similarities between field theories. Having $(CH)_x$ in mind, we include here structural similarities between ϕ^4 and G-N field theories, and with continuum models of Peierls-dimerized chains. ϕ^4-modeling of $(CH)_x$ has been used extensively,[7] but cannot be justified rigorously from the TLM or G-N equations because the effective local potential is strongly nonlinear and nonlocal and perturbation (equivalent to Ginzburg-Landau) expansions are divergent in small amplitude regimes[8] (this difficulty is considerably ameliorated in M-fold commensurate Peierls systems with $M > 2$[9]). Nevertheless, symmetry strongly links the various models and known field theory results help to explain the notable successes of ϕ^4-modeling.[7]

2. SOLITONS IN TRANS-$(CH)_x$: KINKS AND POLARONS.

In the continuum limit,[10] trans-$(CH)_x$ has been described in an adiabatic, mean-field approximation by the effective Hamiltonian[4]

$$H = \int dy \left\{ \frac{\omega_Q^2}{2g^2} \Delta^2(y) + \Psi^+(y) \left[-iv_F \sigma_3 \frac{\partial}{\partial y} + \Delta(y)\sigma_1 \right] \Psi(y) \right\} \qquad (1)$$

where $\Delta(y)$ is the (real) gap or staggered displacement order-parameter, $\Psi(y)$ the (complex) electron field expressed below as a 2-component spinor $\begin{pmatrix} u \\ v \end{pmatrix}$, ω_Q^2/g^2 the net electron-phonon coupling constant, v_F the Fermi velocity, and σ_i the i-th Pauli matrix. Variation of H leads[4] to the single particle

electron wave functions

$$\varepsilon_n U_n(y) = -iv_F \frac{\partial}{\partial y} U_n(y) + \Delta(y) V_n(y)$$
$$\varepsilon_n V_n(y) = +iv_F \frac{\partial}{\partial y} V_n(y) + \Delta(y) U_n(y) \quad , \tag{2}$$

and the self-consistent gap equation

$$\Delta(y) = -g^2 (2\omega_Q^2)^{-1} \sum_{n,s} \left[V_n^*(y) U_n(y) + U_n^*(y) V_n(y) \right] \quad . \tag{3}$$

The summation in (3) is over occupied electron states and s is a spin label (suppressed in (1) and (2)). (2) and (3) are our TLM equations.[4]

It is remarkable that all static solutions to the TLM equations can be found exactly and analytically. As we have stressed, this is due to the precisely soliton[7] properties of the continuum equations.[10] Solutions can be guessed by educated Ansätze or constructed as described in Refs. 2,3. First, the ground state is spontaneously doubly-degenerate with[2-4]

$$\Delta(y) = \pm\Delta_o = \pm W \exp(-\lambda^{-1}) \text{ and } \lambda^{-1} = \pi v_F \omega_Q^2 / g^2 \approx 0.4 \tag{4}$$

in trans-$(CH)_x$. W is the full one-electron bandwidth and $2\Delta_o$ the full bandgap: $\varepsilon_\pm = \varepsilon(k) = \pm (k^2 v_F^2 + \Delta_o^2)^{\frac{1}{2}}$ (see Fig. 1). Explicit electron (plane) wave-functions are given in Refs. 2-4. Second, there are kink defects (solitons)[2-4] interpolating between the degenerate ground states:

$$\Delta_K(y) = \pm \Delta_o \tanh (\Delta_o y / v_F) \quad . \tag{5}$$

The associated electron levels comprise[2-4] extended valence and conduction band states which are distorted (phase-shifted) w.r.t. the defect free case, and an additional localized "mid-gap" state ($\varepsilon_\Gamma = \varepsilon_o = 0$; see Fig. 1). It is worthwhile emphasizing that inhomogeneities in one-dimension will always introduce localized levels within the gap. Kinks may be occupied singly or doubly or unoccupied with the unusual charge assignments summarized in Fig. 1. In all cases (neglecting Coulomb) the kink energy is $E_K = 2\Delta_o/\pi$. Added electrons or holes can stabilize a kink-antikink lattice, also described analytically (as an elliptic function).[4,11] Third are polaron defects[2,3,12] (equivalent to "bags" in field theories), which can be written transparently (c.f. (5)) as

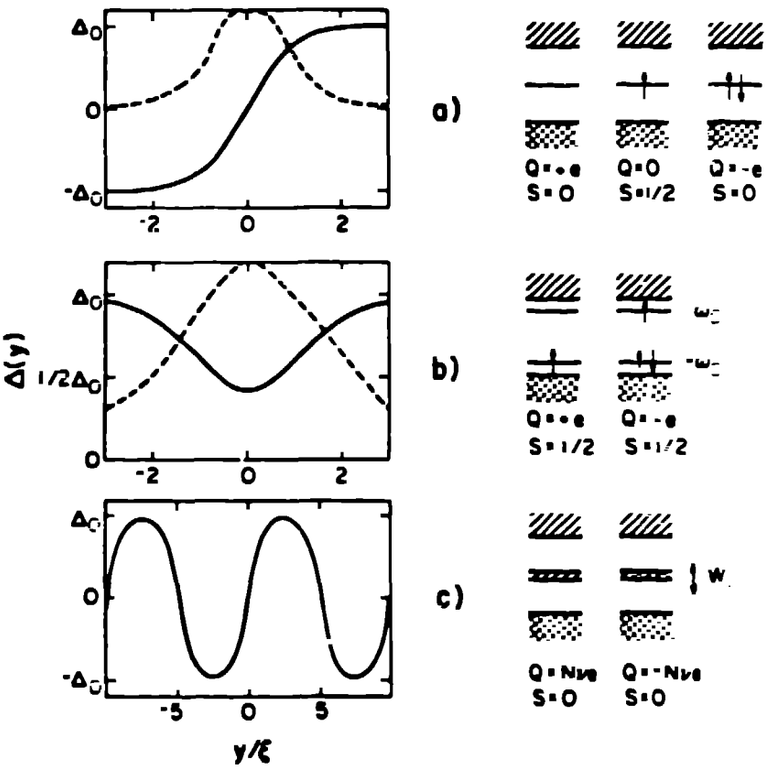

FIGURE 1 Intrinsic defect states in trans-$(CH)_x$ and associated electronic levels (32): a) kink; b) polaron; c) kink lattice. Dashed lines indicate electron densities for localized states (32). $\xi = v_F/\Delta_0$, Q = charge, S = spin, N = number of excess electrons (holes) in the kink lattice[11] and W_1 = localized state bandwidth in this case.

$$\Delta_p(y) = \Delta_o - K_o v_F \left\{ \tanh[K_o(y+y_o)] - \tanh[K_o(y-y_o)] \right\} \quad . \quad (6)$$

Again the extended conduction and valence band electron wave functions are phase-shifted by the defect, but two localized states form symmetrically in the gap (Fig. 1) at $\varepsilon = \pm\omega_o$:

$$\omega_o = (\Delta_o^2 - K_o^2 v_F^2)^{\frac{1}{2}} \; ; \; \tanh(2K_o y_o) = K_o v_F / \Delta_o \quad . \quad (7)$$

It is known[2] (see also energy considerations, §3) that only the singly-occupied (electron or hole) polaron is stable, e.g., double and single occupancy of $\varepsilon = -\omega_o$ and $+\omega_o$, respectively. If the $+\omega_o$ level is unoccupied the polaron collapses to the vacuum, if doubly occupied an infinitely separated charged kink-antikink pair forms. It is important to emphasize that (6) and associated electron wave-functions[2] satisfy (2) for all $K_o(\omega_o)$: $\omega_o \to \Delta_o$ corresponds to uniform dimerization and $\omega_o \to 0$ to infinitely separated kink-antikink. However, the gap equation (3) fixes K_o (c.f. single kink theory[4]). For the interesting singly-occupied polaron (3) gives[2,3,12]

$$\omega_o = K_o v_F = \Delta_o/\sqrt{2} \quad , \quad (8)$$

and this polaron has energy $E_p = 2\sqrt{2}\,\Delta_o/\pi \simeq 0.90\Delta_o$. From (6) and (8) note that this polaron has approximately the same width as the kink (5) ($\simeq$10 A for trans-$(CH)_x$). These are compared in Fig. 1. The polaron is of a strong-coupling variety since polaron binding energy >> maximum phonon energy (the criterion for a valid adiabatic approximation is very similar[1,5]). Furthermore, it is strongly non-perturbative; the perturbative limit (which does not satisfy (3)), $\omega_o \to \Delta_o$, $\Delta_p(y) - \Delta_o \sim \mathrm{sech}^2(K_o y)$, corresponds to a nonlinear Schrödinger description,[2] familiar in one-dimensional polaron theory. The polaron has been observed in independent numerical simulations[13] of the lattice model[5] of $(CH)_x$ and agrees well with (6)-(8). Interestingly, for spinless fermions[14] no stable polaron is found numerically[15] in agreement with the relevant N = 1 Gross-Neveu model results.[2,3]

3. SOLITONS IN CIS-$(CH)_x$: POLARONS.

The notion of a degenerate ground state in Peierls distorted electron-phonon systems, leading as it does to kinks with fractional charge,[1] is of wider application than trans-$(CH)_x$, especially when extended to M-fold commensurate systems[4] (TTF-TCNQ under pressure, monoclinic TaS_3, etc.).

However, in another sense polarons are likely to be more ubiquituous than kinks in a general scenario of polymers since they do not require conformation degeneracy (or indeed multi-conformation structures at all). Many polymers, unlike trans-$(CH)_x$, have a nondegenerate ground state with additional conformations at higher energy; e.g., cis-$(CH)_x$, polydiacetylenes,[15] polyphenylenes,[13] etc. Energy differences and barriers between local minima may each be small or large, but (within steric constraints) inhomogeneous states are again possible. However, static kink states are now not possible unless pinned by impurities, chain ends, etc. (See Fig. 2.) In equilibrium on the pure chain only confined kink-antikink pairs (i.e., polarons) are possible. The notion of a confinement energy is shown in Fig. 2. Since interesting doping, ESR observations etc, are by no means limited to trans-$(CH)_x$, polydiacetylenes,[15] polyphenylenes,[13] etc. Energy differences and barriers between local minima may each be small or large, but (within steric constraints) inhomogeneous states are again possible. However, static kink states are now not possible unless pinned by impurities, chain ends, etc. (See Fig. 2.) In equilibrium on the pure chain only confined kink-antikink pairs (i.e., polarons) are possible. The notion of a confinement energy is shown in Fig. 2. Since interesting doping, ESR observations etc., are by no means limited to trans-$(CH)_x$, it is important to understand a wider spectrum of polymers.

On these general grounds, polarons (singly or doubly occupied) are expected very generally in the absence of degeneracy. The following model (due to Brazovskii and Kirova[12]) is especially valuable since it again allows an explicit solution as for trans-$(CH)_x$. We assume a gap parameter $\tilde{\Delta}(y) = \Delta(y)+\Delta_e$, where $\Delta(y)$ is sensitive to electron feedback (as in §2) but Δ_e is a constant extrinsic component. Such an Ansatz is quite plausible for, say, cis-$(CH)_x$ in terms of σ-bond effects on distortions. In any event it immediately yields a soluble field theory (continuum limit) as for the degenerate case of §2. We merely replace $\Delta(y)$ with $\tilde{\Delta}(y)$ in the Dirac (electron) equations (2), but not in the corresponding gap equation (3) where Δ_e does not enter. Thus, the structure of the electron spectrum and eigenfunctions is precisely that of §2, but the new gap equation leads to a modified renormalization condition and constraints on bound state eigenvalues. Specifically, we find after some algebra that the ground state $\tilde{\Delta}(y) = \tilde{\Delta}_o = \Delta_o + \Delta_e$ is given by

$$\Delta_e = \lambda\tilde{\Delta}_o \ln(\tilde{\Delta}_o/\Delta_o) \quad ; \quad \Delta_o = W \exp(-\lambda^{-1}) \quad . \tag{9}$$

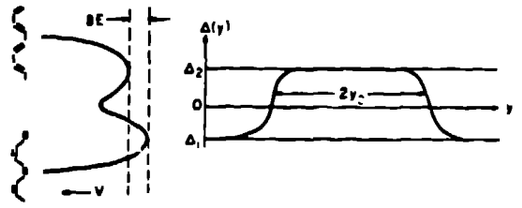

FIGURE 2 Illustration of "confinement" (see §3) for non-degenerate conformations, e.g. (for cis-$(CH)_x$) cis-transoid (metastable) and trans-cisoid (stable), leading to polarons and bi-polarons (c.f. Ref. 23 but note the extended forms expected (§3)). Recall also metastable state decay and nucleation theories, which have potential polymer applications.

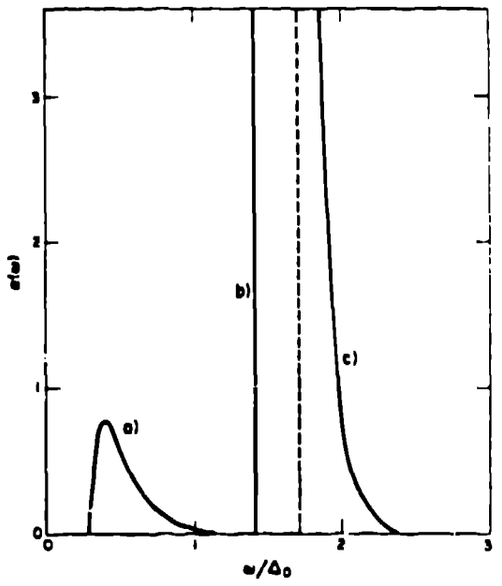

FIGURE 3 Optical absorption (α) due to (electron) polaron. Contributions a), b), c) correspond to α_3, α_1, α_2, respectively (§4).

Note that W and λ are not in general the same as for trans-$(CH)_x$ in §2. As stated, single kinks no longer satisfy the gap equation, but polarons are again given by (6,7) (with Δ_p, $\Delta_o \to \tilde{\Delta}_p$, $\tilde{\Delta}_o$ and the same valence and conduction band electron functions). The only difference is the location of ω_o, which we find is now determined from

$$\gamma K_o v_F = \frac{1}{4}\pi\omega_o(n_+ - n_-) + \omega_o \tan^{-1}(\omega_o/K_o v_F) \quad . \tag{10}$$

Here n_+, n_- denote the occupancies of $\varepsilon_\pm = \pm\omega_o$ (Fig. 1), respectively, and $\gamma = \Delta_e/\lambda\tilde{\Delta}_o$. For $\gamma = 0$, we recover the results of §2: $n_+ = 2$, $n_- = 2$ gives $\omega_o = 2$ gives $\omega_o = 0$, (infinitely separated kinks); $n_+ = 1$, $n_- = 2$ gives (8). For $\gamma > 0$, (10) gives nontrivial results for either single or double (electron or hole) occupancy, as anticipated (Fig. 2). The putative kink separation (i.e., polaron width, c.f. (6,7)) $2y_o = K_o^{-1} \tanh^{-1}(K_o v_F/\tilde{\Delta}_o)$ increases with occupation, and correspondingly the polaron becomes shallower. [This general polaron trend is observed in discrete simulations of polyparaphenylene[13] (in which benzene and quinolene ring structures are nondegenerate).] The same conclusions follow from energy calculations for the general profile (6) and then minimization w.r.t. $\omega_o y$ (or K_o or y_o) for each occupation. In such calculations, phase-shift effects on the extended electron state due to the inhomogeneities must be carefully included, but this is by now very familiar in soliton physics.[16] We find generally[12]

$$E_p[\tilde{\Delta}_p(\tilde{\omega}_o)] - E_o[\Delta_o] = (n_+ - n_- + 2)\omega_o + \frac{4}{\pi} K_o v_F - \frac{4}{\pi}\omega_o \tan^{-1}(K_o v_F/\omega_o) + \frac{4}{\pi}\tilde{\Delta}_o\gamma[\tanh^{-1}(K_o v_F/\tilde{\Delta}_o) - K_o v_F/\tilde{\Delta}_o] \quad , \tag{11}$$

from which all our previous results follow for $\gamma = 0$ or > 0. In particular note that the last term in (11) is essentially the confinement energy (Fig. 2) $\to (4\tilde{\Delta}_o/\pi\lambda v_F)\Delta_e(2y_o)$, for $y_o >> v_F/\tilde{\Delta}_o$. The Ansatz (6) and corresponding energy (11) are probably also the best single-parameter way of studying all possible kink-antikink interaction energies (in the continuum limit). For instance with $n_+ = n_-$ and $\gamma = 0$, (11) gives $(E_p(y_o) - E_p(\infty))/E_p(\infty) \to \exp(-2y_o/\zeta)$, $y_o >> \zeta$, where $\zeta = K_o^{-1} = (v_F/\Delta_o)(\tanh 2K_o y_o)^{-1}$. This and all other kink-antikink predictions are in good agreement with discrete simulations,[13,17] but we emphasize that these (and (6)) are static descriptions and do not presage the complex dynamics expected in collision processes[2,13] (See §5).

4. OPTICAL ABSORPTION FROM POLARONS.

The influence of a single kink and the kink-lattice on the optical absorption have been studied in detail.[18] Here we concentrate on the contribution of a single polaron in *trans*-$(CH)_x$. Due to the symmetry of the TLM-equations (2), the transition from the valence band (v.b.) to the singly occupied localized state at $\varepsilon = +\omega_0$ (See Fig. 1) is the same as the transition from the doubly occupied state at $\varepsilon = -\omega_0$ to the conduction band (c.b.). The same holds true for the transitions v.b. $\to -\omega_0$ and $+\omega_0 \to$ c.b.. Using the wave functions u, v in the presence of a polaron,[2] the optical absorption $\alpha(\omega)$ involving transitions to or from a localized state can be calculated; the results are shown in Fig. 3. (The detailed formulae will be given elsewhere.[19])

The transition $-\omega_0 \to +\omega_0$ ($\alpha_1(\omega)$) results in a δ-peak at $\omega = 2\omega_0$; v.b. $\to +\omega_0$ ($\alpha_2(\omega)$) is singular at

$$\omega = \left(1 + \frac{1}{\sqrt{2}}\right)\Delta_0 ; \quad \alpha_2(\omega) \sim \left(\left(\frac{\omega-\omega_0}{\Delta_0}\right)^2 - 1\right)^{-\frac{1}{2}} ,$$

whereas the transition $+\omega_0 \to$ c.b. ($\alpha_3(\omega)$) does not show any singularity. Due to the accessible spin states, the total contribution of a single (electron) polaron to the optical absorption is $\alpha_P(\omega) = \alpha_1(\omega) + \alpha_3(\omega) + 3\alpha_2(\omega)$ with a total intensity of

$$\int \alpha_P(\omega)d\omega = 4.17 e^2 v_F \frac{2\pi}{L} \frac{v_F}{\Delta_0}$$

which is greater by a factor of 3.66 than the corresponding results for a single kink.[18] A full account of these results together with the interband contribution for *trans* as well as for *cis*-$(CH)_x$ will be given elsewhere.[19]

5. $(CH)_x$ AND ϕ^4 PHENOMENOLOGY.

We remarked in §1 on the formal failure of Landau-Ginzburg-expansion-motivated ϕ^4 modeling[7,8] of half-filled band Peierls chains in small amplitude order parameter (Δ) regimes. Nevertheless, the qualitative successes are very real and it is worthwhile emphasizing the structural relationships between these problems (and G-N) which are known from field theory studies. We summarize some of these briefly:[2]

(i) The prototype ϕ^4 Lagrangian

$$\mathcal{L} = \frac{1}{2}(\partial_\mu \phi \partial^\mu \phi) - \frac{1}{4}\lambda(\phi^2 - f^2)^2 \qquad (12)$$

(f, λ constants) has the desired doubly degenerate ground state but this arises from explicit spontaneous symmetry

breaking. In G-N or $(CH)_x$ the effective potential for the boson field ($\sim\Delta_o^2 \ln\Delta_o^2$) arises dynamically. In a sense the ϕ^4 potential imitates valence band electron effects in $(CH)_x$.
(ii) Unlike (12) there are no spatial gradients in G-N or continuum models of $(CH)_x$ to control characteristic inhomogeneity size scales. Their role is played by nonlocalities coming from the valence band.
(iii) All three models support kinks (because of the common effective potential topology (i)), kink lattice (as elliptic functions), polarons and "breathers." However, to parody the associated electronic properties in $(CH)_x$ or G-N, it is necessary to explicitly couple fermions to the ϕ-field. This can be achieved in several ways (some allowing analytic progress)[2] which do indeed mirror the G-N bound state structures and spin/charge assignments.
(iv) Dynamics most clearly distinguish the models (see also §6), but are also the least well studied features. Semiclassicial G-N enjoys literal soliton (i.e., purely phase-shifting) collisions.[2,3] This is not expected for $(CH)_x$. Indeed the adiabatic ion dynamics usually assumed[4,5] in $(CH)_x$ is rather that of ϕ^4 (12). Collisions between ϕ^4 kinks (and breathers) have a fascinating and complex structure, much of which is due to a kink shape oscillation mode.[20] A similar mode has recently been predicted[21] in continuum $(CH)_x$ theory, so that similar collision structure might be anticipated. This is somewhat supported by numerical simulations.[13]

6. DISCUSSION.

Studies are on-going in two major directions. First to clarify those problems where field theory connections are not available or are misleading, and second to assess impacts on experiments.

The difference in dynamics between continuum $(CH)_x$ and G-N is of central importance. We have already mentioned the structureless collisions in G-N (§5). It is also worthwhile emphasizing that the band of "breather" states[3] in G-N (of which the static polaron is the lowest member) can be thought of as a conventional polaron band,[2] but with real $(CH)_x$ dynamics shake-off phonon structure and decay channels can be expected. A second role for dynamics is to determine the quantized spectra of the different models. The full G-N S-matrix is known reasonably conclusively (See Ref. 2) and for N = 2 only kinks survive quantum fluctuations. However, our $(CH)_x$ mapping was to massive (i.e., static) G-N. Preliminary quantum Monte Carlo studies[22] show that quantum fluctuations are still large with $(CH)_x$ dynamics but on the

other hand first order quantum corrections[21] do not appear to destabilize the polaron. In addition, it is crucial to assess interchain coupling and e-e interaction effects. (The latter can produce exciton-polarons, recombination barriers, etc.)

Turning to experimental implications of polarons (and more generally breathers) we note first that on energy grounds[2,12] (§§2,3) single electrons added to cis-or trans-$(CH)_x$ or similar polymers (by doping, injection, etc.) should form polarons initially and (with additional electrons) a kink (trans) or doubly occupied polaron (bi-polaron) (cis) lattice. (Note also that a neutral kink and singly-charged polaron (trans) will produce a charged kink.) Timescales will depend on pinning mechanisms, Coulomb recombination barriers, etc. It is clearly necessary to settle uncertainties, over isomerization of trans from cis upon doping.[23] If polarons can be induced then their optical absorption (§4) is sufficiently different from kinks for experiments to be encouraged and predictions of "mid-gap" and IR absorption compared. A plausible scenario has been given for[24] the contrasts between cis- and trans-$(CH)_x$ upon photoinjection, particularly regarding observation (or not) of a photocurrent and luminescence. The same scenario should include other conjugated polymers; polyphenylenes, polydiacetylenes, etc. It is clear, however, that for quantitative predictions more detailed understanding of kink/polaron collisions are needed; the long carrier lifetimes (~300 sec.) implied in trans data[24] already suggests complex dynamics (as in ϕ^4), more generally, as in conventional small polaron semiconductor photoinjection theory,[25] explanation of photocurrent and luminescence needs details of carrier transport, trap times, recombination kinetics. Even when confinement energy encourages kink-antikink recombination (e.g., cis-$(CH)_x$), this may proceed through complex intermediate (e.g., breather) metastable states because of intrinsic nonlinear dynamics. Here again we also need to understand e-e interaction effects on defect states--they can in general lead to "exciton-polarons" and recombination barriers. Polaron transport theory has been described by Holstein in these Proceedings. Tunnel (e.g., diode) injection is in principle a good way to observe polaron band and shake-off phonon structures, although energy changes are rather small in the case of $(CH)_x$ (§§2,3). It is well to recall the role of polarons expected on the metallic side of the doping transition as in conventional amorphous semiconductor theory. Indeed, finally, we express the hope that

$(CH)_x$ and general amorphous semi-conductor theories will move closer with concepts of dangling bonds, polarons bipolarons, negative U states, pseudogap, photoinjected carrier kinetics and recombination time scales, etc.. This would be to their mutual benefit.

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